

Final Report, N00014-97-1-0745
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Electron Energy Levels in Magnetic Nanoparticles

Work under this grant progressed in two primary directions: (1) Measuring electron tunneling via individual "electron-in-a-box" quantum energy levels inside magnetic particles, and subsequently studying the energy spectra to determine the effects of magnetic exchange interactions and magnetic anisotropy forces, and (2) Pursuing a discovery made in work funded by this grant that spin-polarized currents flowing in magnetic multilayers can cause magnetic switching in nm-scale devices.

Tunneling via individual electronic states in cobalt nanoparticles (work performed by postdoc Sophie Guéron and graduate student Mandar Deshmukh, publications #2, 3, and 8).

The need for a detailed understanding of the electronic states in small ferromagnetic elements is motivated by the rapid rate of miniaturization that is proceeding in magnetic storage technologies. Under this grant, my group made the first measurements of the energy spectrum of discrete quantum states inside single-domain ferromagnetic cobalt particles 1-4 nm in diameter. We gained new insights into the nature of electron tunneling in small ferromagnets -- namely that collective spin-wave excitations mix into the spectrum so that simple independent-electron models do not provide an accurate description. We also measured the changes in the electronic energy levels as a function of magnetic field. As an applied field causes the magnetic moment of the nanoparticle to rotate and then reverse, the energy of each quantum state evolves and then undergoes an abrupt jump. An analysis indicates that these effects can be understood as a consequence of magnetic anisotropy within the particle, but that the magnitude of this anisotropy has large fluctuations (tens of percent) with the addition even of a single electron to a nanoparticle. Dealing with these fluctuations will be a significant challenge if nm-scale magnetic particles are incorporated into non-volatile magnetic memory technologies.

Fabricating tunnel barriers: In order to perform tunneling spectroscopy of individual magnetic nanoparticles, it was necessary to develop a means of forming high-quality tunnel barriers on metal nanoparticles using materials other than the native oxide of the metal. We succeeded in doing this for cobalt particles using two different techniques. First we adapted the procedure used by the MIT and IBM groups (among others) for making magnetic tunnel junctions -- depositing at liquid nitrogen temperature a thin film of aluminum on the magnetic material, and then oxidizing the aluminum. Second, we learned that we could make excellent tunnel junctions by directly depositing a thin film of aluminum oxide on the particles. Both techniques produce similar results for the discrete tunneling spectra. The second technique generally does not work for larger area tunnel junctions due to pinholes. However, since our tunnel junctions are only a few square nm in area, we can have a high yield of pinhole-free devices even with an average pinhole density large enough to destroy the function of larger-area junctions. The method of direct deposition of aluminum oxide is now our preferred technique, so as

to avoid ambiguities in experimental interpretation that might come from the presence of un-oxidized aluminum. This method should be widely applicable to other metals.

Energy-level spacing: Two results from our studies of Co nanoparticles indicate that simple two-current models commonly used to understand the physics of larger ferromagnetic tunneling devices become inapplicable on the nanometer scale. By two-current models we mean that current flow is assumed to be due to independent spin-up and spin-down electrons, which may have different tunneling densities of states near the Fermi level. The first experimental result in conflict with this picture is that the energy-density of tunneling resonances is at least an order of magnitude greater than can be explained by independent-electron states within the nanoparticle, even taking into account the large density of d-band states near the Fermi level in cobalt. The spacing is, instead, comparable to the energy of the spin-wave excitations of the particle's magnetic moment. This suggests that accurate models of tunneling in magnetic nanoparticles must take into account not only the single-electron-type tunneling excitations but also how single-electron excitations mix with the low-energy *collective* excitations of the magnetism. The second experimental result in conflict with simple models is that we do not observe comparable tunnel coupling to the electrodes for spin-up and spin-down energy states, with just a difference in the density of states (for Co, the expected polarization is about 35%). Even when we observe 10 or more resonances in a given device, they all shift as a function of magnetic field with the same sign of slope, indicating that they all have the same spin orientation instead of some being spin-up and others spin-down. We are still in the midst of investigating this effect, but one possible explanation at this time is that spin-orbit scattering serves to mix the spin-up and spin-down quantum states.

Magnetic response of individual states: We have also observed that there is a strong coupling between the direction of the magnetic moment vector for the nanoparticle and the energy of each of the electron-in-a-box states. When a small magnetic field is applied to reorient the magnet moment, the levels shift. This is a very different effect than changes in the total magnetization previously monitored by SQUID techniques or MFM. The total magnetization can be thought of as the derivative with respect to a magnetic field of the *total* energy of all the electrons in the particle: $M = dE_{\text{tot}}/dH$. In contrast, we are able to measure the magnetic response of the tunneling resonances for *individual* electronic states. Over the field range for which the moment is expected to turn continuously between the easy-axis direction and the direction of the applied field, our levels shift continuously, but for a reversed field that is sufficiently large, all the levels for a given particle jump discontinuously at the same field value. We interpret this as the field where the particle's magnetic moment undergoes reversal. We have also made progress on a theoretical understanding of the nature and size of this coupling, with a simple model that calculates the contribution of magnetic anisotropy and the Zeeman energy to the total energy of the many-electron eigenstates within the nanoparticle. The detailed magnetic-field dependence indicates that the magnetic anisotropy energy varies between different states on the same particle, by tens of percent, even if the states differ only by the addition of one electron. These intrinsic, mesoscopic fluctuations may explain previous observations by Chris Murray at IBM, Yorktown Heights, that size-

selected cobalt nanoparticles formed by chemical means have switching fields that vary by 10% or more.

Current work: Work on this project will be continuing, under funding from the NSF. The current goals are to understand the processes of spin-dependent tunneling between a ferromagnetic electrode and the quantum states in a magnetic nanoparticle, to use the sensitivity of the discrete energies to the direction of the particle's magnetic moment vector to study the switching dynamics of the moment in real time, and to extend the work to other materials, such as copper, gold (publication #7), and palladium, which some theories predict should be slightly magnetic in nanoparticle form.

Exchange-force-mediated switching of domains in magnetic multilayers by spin-polarized currents (work performed by graduate student Ed Myers, publications #1, 4, 5, and 6).

Following a suggestion by the theorist John Slonczewski, we examined the effects of currents flowing perpendicularly through samples consisting of Co/Cu/Co metallic layers adjacent to a point-contact, made using the same silicon-nitride nano-holes employed in the nanoparticle tunneling devices. We were able to verify the prediction that the spin-polarized currents flowing in such a geometry can apply torques to the magnetic layers sufficient to cause their magnetic moments to reorient, due to a new mechanism based on local exchange interactions and not having anything to do with a magnetic field. The new mechanism is actually a simple (but previously unappreciated) consequence of Newton's third law. When a current of electrons is incident on a thin magnetic thin film, the film acts as a spin-filter, allowing (for Cu/Co interfaces) preferential transmission of majority-spin electrons. When a previously-spin polarized current is incident on a magnetic layer, the spin-filtering can result in a deposition of spin angular momentum into this magnetic layer, or in other words, a torque is applied to the magnetic moment of layer. We demonstrated that this torque can be employed in a useful way to provide the means for a simple magnetic memory element -- we made a device in which two magnetic layers could be switched to a parallel orientation by a current pulse flowing in one orientation, and then switched anti-parallel by a current pulse directed in reverse. This exchange-induced switching is 100 times more powerful for a given amount of current than the Oersted-Maxwell magnetic field generated by the current, and it is local rather than being long-ranged, so that it may be a useful mechanism in improving the design of high-density magnetic random access memory (MRAM).

Since our initial demonstration, I have also worked with Jordan Katine and Bob Buhrman to demonstrate the effect in a more practical geometry -- a Co/Cu/Co pillar approximately 100 nm wide. Work is underway to measure the switching speed, and to optimize the materials and device geometries for utilizing this effect. The future work will be supported in part by the MRSEC program of the NSF, and I am also seeking support from the DARPA SPINs program.

Human Resources Development: This grant provided full or partial support (in conjunction with an NSF grant) for 1 postdoc [Sophie Guéron], 2 graduate students [Ed Myers and Mandar Deshmukh], and 2 undergraduates [Stephen Meyer (a Cornell student) and Matthew Dearing (a summer student from Illinois Wesleyan)]. Dr. Guéron has

moved on to her first-choice job, as a professor with the CNRS in France, at the Université Paris Sud. Myers and Deshmukh are still with my group. Two other graduate students worked on projects related to the grant, but were supported by other funds. These are David Salinas (who was recruited away by McKinsey Consulting before finishing his Ph.D.) and Jason Petta (currently in my group). I have given seminar presentations on this research to undergraduates in Cornell's experimental laboratory course.

Publications

1. "Current-Induced Switching of Domains in Magnetic Multilayer Devices"
E. B. Myers, D. C. Ralph, J. A. Katine, R. N. Louie, and R. A. Buhrman
Science **285**, 867 (1999).
2. "Tunneling via Individual Electronic States in Ferromagnetic Nanoparticles"
S. Guéron, Mandar M. Deshmukh, E. B. Myers, and D. C. Ralph
Phys. Rev. Lett. **83**, 4148 (1999). (cond-mat/9904248)
3. "Electron Energy Levels in Superconducting and Magnetic Nanoparticles"
D. C. Ralph, S. Guéron, C. T. Black, and M. Tinkham
Physica B **280**, 420 (2000).
4. "Current-Driven Magnetization Reversal and Spin Wave Excitations in Co/Cu/Co Pillars"
J. A. Katine, F. J. Albert, R. A. Buhrman, E. B. Myers, and D. C. Ralph
Phys. Rev. Lett. **84**, 3149 (2000). (cond-mat/9908231)
5. "Point-Contact Studies of Current-Controlled Domain Switching in Magnetic Multilayers"
E. B. Myers, D. C. Ralph, J. A. Katine, F. J. Albert, and R. A. Buhrman
J. Appl. Phys. **87**, 5502 (2000).
6. "The Role of Spin-Dependent Interface Scattering in Generating Current-Induced Torques in Magnetic Multilayers"
Xavier Waintal, Edward B. Myers, Piet W. Brouwer, and D. C. Ralph
submitted to *Phys. Rev. B*, cond-mat/0005251
7. "Measurements of Discrete Electronic States in a Gold Nanoparticle Using Tunnel Junctions Formed from Self-Assembled Monolayers"
Jason R. Petta, D. G. Salinas, and D. C. Ralph
submitted to *Applied Physics Letters*
8. "Spectroscopy of Discrete Energy Levels in Ultrasmall Metallic Grains"
Jan von Delft and D. C. Ralph
to appear in *Physics Reports*

Invited Talks at Conferences

1. "Interacting Electrons in a Box -- Tunneling Measurements of Electronic Energy Levels in Single Metal Particles,"
Adriatico Research Conference -- STM-Based Lithography and Atomic Electronics
July 15-18, 1997, Trieste, Italy.
2. "Effects of Electron Interactions on the Tunneling Spectra of Aluminum Nanoparticles"
Israeli Science Foundation Workshop -- Strong Interactions in Quantum Dots,
October 25-30, 1997, Dead Sea, Israel.
3. Lecturer in the International Summer School in Nanophysics (3 Lectures)
August 11-14, 1998, Helsinki University of Technology, Otaniemi, Finland.
4. "Issues of Magnetism and Spin-Orbit Scattering in Quantum Dots"
Extended Research Workshop on Disorder, Chaos, and Interaction in Mesoscopic Systems
August 17-20, 1998, Trieste, Italy.
5. "Electron Energy Levels in Superconducting and Magnetic Nanoparticles"
22nd International Conference of Low Temperature Physics
August 4-11, 1999, Helsinki, Finland
6. "Electron tunneling via single energy levels in cobalt nanoparticles"
Gordon Research Conference on Magnetic Nanostructures
Feb. 13-17, 2000, Ventura, CA
7. "Current-Controlled Domain Switching in Magnetic Multilayers By Means of Spin Transfer"
Spring Materials Research Society Meeting
April 24-28, 2000, San Francisco, CA
8. "Interacting-Electron States inside Ferromagnetic and Superconducting Particles"
Workshop -- Interaction and Chaos in Mesoscopic Systems
May 12-14, 2000, Theoretical Physics Institute, University of Minnesota
9. "Effects of Interactions, Chaos, and Non-Equilibrium on Electron Tunneling in nm-Scale Metal Particles"
Nobel Symposium, Quantum Chaos Y2K
June 13-17, 2000, Bäckaskogs Castle, Sweden
10. "Torques and Electron Tunneling in Nano-Magnets"
Gordon Conference on Correlated Electron Systems
June 25-30, 2000, Plymouth State College, NH

11. Lecturer at the United States Summer School in Condensed Matter and Materials Physics: Introduction to Superconductivity -- Fundamentals and Applications
- Lecture 1: Thinking about Interacting Electrons in Systems with Discrete Energy Levels
 - Lecture 2: Superconductivity from the Micron to the Nanometer Scale
 - Lecture 3: The Destructive Effect of a Magnetic Field -- Orbital vs. Spin Pair Breaking
- July 10-14, 2000, Boulder, CO

Talks at Universities and Laboratories

- "Interacting Electrons in a Box -- Measurements of Electronic Energy Levels in Single Metal Particles"
- University of Virginia, Sept. 26, 1997, Physics Colloquium
 - National High Magnetic Field Laboratory, Tallahassee, FL, Oct. 10, 1997, Condensed Matter Sem.
 - North Carolina State University, Raleigh, NC, Feb. 16, 1998, Physics Colloquium
 - Michigan State University, East Lansing, MI, April 20, 1998, Condensed Matter Seminar
 - Princeton University, Princeton, NJ, April 27, 1998, Electronic Materials and Devices / ISS Joint Seminar
 - IBM, Yorktown Heights, NY, Oct. 21, 1998, Nanostructures Seminar
 - University of Minnesota, Minneapolis, MN, March 3, 1999, Physics Department Colloquium
- "Nano-Magnetism"
- University of Rochester, Rochester, NY, April 19, 1999, Solid State Seminar
- "Torques and Tunneling in Nanomagnets"
- Cornell University, Sept. 23, 1999, Materials Science and Engineering Seminar
 - Yale University, New Haven, CT, Nov. 5, 1999, Condensed Matter Seminar
 - Stanford University, Stanford, CA, Nov. 18, 1999, Condensed Matter Seminar
 - Duke University, Durham, NC, Nov. 29, 1999, Condensed Matter Seminar
 - IBM, Yorktown Heights, NY, Jan. 20, 2000, Physical Sciences Seminar
 - Georgetown University, Washington DC, April 13, 2000, Physics Colloquium
- "Spin-tronics -- Electronics that Uses the Electron's Spin"
- Colgate University, Hamilton, NY, April 18, 2000, Physics Colloquium
- "Manipulating Nanomagnets with Spin-Polarized Currents"
- NIST, Boulder, July 11, 2000, Condensed Matter Seminar

Presentations by Members of My Group

Invited Talks at Conferences

E. B. Myers, "Magnetic Domain Switching Induced by Spin-Transfer from Spin-Polarized Currents"
2000 March Meeting of the American Physical Society
March 20-24, 2000, Minneapolis, MN.

Seminars at Universities and Laboratories

S. Guéron, "Electronic states in nanometer-scale superconductors and magnets"
University of Illinois, Champaign-Urbana, Oct. 22, 1998
Rutgers University, Nov. 10, 1998
Orsay, France, Jan. 5, 1999

S. Guéron, "Single-Electron Tunneling in Cobalt Nanoparticles"
Yale University, April 7, 1999.
UCLA, Los Angeles, CA, Nov. 10, 1999
UC San Diego, San Diego, CA, Nov. 12, 1999

Contributed Presentations

"Single Electron Tunneling Through Magnetic Nanoparticles", S. Guéron and D. C. Ralph, contributed talk, 1999 APS March Meeting, Atlanta, GA, March 24, 1999.

"Asymmetric Current-Induced Transitions in Single Magnetic Interface Point Contacts", E. B. Myers, D. C. Ralph, and R. A. Buhrman, contributed talk, 1999 APS March Meeting, Atlanta, GA, March 24, 1999.

"Point Contact Studies of Current-Controlled Domain Switching in Magnetic Multilayers", E. B. Myers, contributed talk at the 1999 Magnetism and Magnetic Materials conference, San Jose, CA, Nov. 14-18, 1999.

"Electron Tunneling via Individual Energy Levels in Cobalt Nanoparticles", S. Guéron, contributed poster at the 1999 Magnetism and Magnetic Materials conference, San Jose, CA, Nov. 14-18, 1999.

"Measurement of discrete electronic states in a gold nanoparticle using tunnel junctions formed from self-assembled monolayers", J. Petta, D. C. Ralph, and C. B. Murray, contributed talk, 2000 APS March Meeting, Minneapolis, MN, March 20-24, 2000.

"Tunneling spectroscopy of ferromagnetic nanoparticles," M. M. Deshmukh, S. Guéron, E. B. Myers, J. Petta, and A. Pasupathy, contributed talk, 2000 APS March Meeting, Minneapolis, MN, March 20-24, 2000.